Preprints of papers presented at the
Fourth International Conference on Methods and Applications of
Radioanalytical Chemistry

and

submitted for publication in the
Journal of Radioanalytical and Nuclear Chemistry

Classification of Alpha-active Workplace Aerosols Based on Coefficient of Transportability as Measured by the Dialysis Method

V. F. Khokhryakov[1], K. G. Suslova[1], I. A. Tseveloyova[1], E. E. Aladova[1], R. E. Filipy[2]

[1]Branch No. 1, Federal Research Center Biophysics Institute, Ozyorsk Road 19, Ozyorsk, Chelyabinsk Region, Russia 456780.

[2]U. S. Transuranium and Uranium Registries, Washington State University, 100 Sprout Road, Richland, WA 99352.

Corresponding Author: R. E. Filipy

ABSTRACT

This report describes a method by which potentially inhaled workplace aerosols containing plutonium compounds are classified on the basis of measured transportability in Ringer's solution. It is suggested that the criterion "transportability" be used in the ICRP respiratory tract model. Transportability is measured as the fraction of plutonium alpha activity, deposited on a collecting filter, that passes through a semi-permeable membrane in Ringer's physiological solution during two days of dialysis. First order kinetic equations are used for explanation of dialysis results. The dissolution characteristics of alpha-active aerosols are important in interpretation of their passage from the lungs after inhalation.

INTRODUCTION

The retention and clearance of inhaled Pu aerosols deposited in a worker's lungs are determined in part by the particle size distribution and their solubility [1,2]. Experimental studies with animals [3,4] and available information on deposition in worker's body after accidental inhalation exposures [5,6] show that the lung clearance of plutonium particles varies widely depending on chemical form of the inhaled compounds. The chemical properties of actinides, in particular of plutonium, have been studied in detail; however, the data about their solubility in the classic chemical sense are not useful for predicting their behavior in the body.

Due to absence of strict correlation between metabolic parameters and chemical forms of radionuclide compounds, the ICRP dosimetry models of the respiratory tract group radioactive aerosols into three classes, D, W, or Y, i.e.: those with rapid, moderate and slow removal from the respiratory tract [1,2]. Because of the lack of data describing the correlation between lung clearance rates and physico-chemical properties of inhaled aerosols, the indicated classification has a qualitative character. Also, workplace aerosols generally contain not one, but a mixture of several chemical compounds. And

last, the rate of lung clearance and the capacity to pass through the semi-permeable membranes in the body are influenced by particle size.

A number of investigators studied tests for aerosol characteristics, which would allow prediction of biological behavior of inhaled radionuclides in man [7-9]. However, the literature data does not reveal criteria for characterization of industrial aerosols, that can be used for practical dosimetry assessments. We have performed a detailed study of the dissolution characteristics of plutonium aerosols sampled at plutonium-handling facilities of the Mayak Industrial Association [10].

This report contains a description of a dialysis method for classification of workplace plutonium-containing aerosols on the basis of in vitro solubility for worker dosimetry assessments.

Materials and Methods

Aerosol samples for dissolution studies were collected by standard aspiration methods on AFA-RSP-20 filters from different workplaces of uranium and plutonium reprocessing plants. Each filter was sandwiched between two membrane filters (type Vladipor with pore size 0,15-0,25 μm) and mounted in a specially made holder. This assembly (Fig. 1) was placed in a 300-ml glass beaker with 150ml volume of Ringer's

physiological solution at room temperature without stirring. At intervals of 1, 3, 7, 15, and 24 h after the beginning and at 24-h intervals thereafter, the solvent was removed and replaced with a fresh solution, and the plutonium activity was determined in the removed solution. At the end of 14 d the plutonium remaining on the filter was assayed and the activity value was added to those of the solutions to determine the amount of plutonium initially present on the filter. Samples were analyzed using methods described in a previous report [11]. The dissolution rate was expressed as the percent of plutonium dissolved from the total initial content.

Results

In order to evaluate the dialysis method for the purpose of dosimetry classification, the dialysis of plutonium citrate and plutonium polymeric nitrate was studied. The dissolution rate of Pu citrate was much higher than that for polymeric Pu with $84.7 \pm 9.7\%$ of Pu citrate dissolved from the initial amount after two days of dialysis. The polymeric Pu was less soluble then the citrate with only $4.4 \pm 1.7\%$ of Pu in the solution. Pu citrate is a highly stable complex that is referred to monomeric plutonium; it has a high capacity for diffusion through a semi-permeable membrane during the dialysis. The low dissolution rate of polymeric plutonium can be explained by the larger particles size relative to the membrane pore size of 0.25 μm. Another reason for slow dialysis of polymeric plutonium may be adsorption on the membrane.

Appreciable differences in the dialysis of the two plutonium compounds were recorded and a good correlation between the dialysis rate and compound chemical form was noted.

The composition of workplace aeorsols may be very complex. As a rule, these materials contain a mixture of plutonium compounds of varied chemical forms depending on the technological process in the workplace where the aerosols were generated. Therefore, during dialysis of workplace aerosols in the Ringer's solution, many different interactions can occur.

Analysis of dialysis kinetics of a large number of plutonium-containing aerosols revealed biphasic rate profiles. In all cases, there was fast dissolution in the earlier stage which could last from several hours to one day for aerosols sampled at different workplaces. After 1 day, the dissolution rate of plutonium was reduced by two or more orders of magnitude relative to the value obtained for the first several hours and then the rate became nearly constant. The results of these studies are demonstrated by dialysis of typical plutonium aerosols sampled from two different plutonium-processing areas (Fig. 2).

The presence of two phases in the first approach can be described by a system of first order equations as follows:

$$dQ_1/dt = - \lambda_1 Q_1 + \lambda_2 Q_2 \qquad (1)$$

$$dQ_2/dt = - \lambda_2 Q_2 \qquad (2)$$

Where:

Q_1 is the fraction of rapidly dissolving radionuclide at time, t,

Q_2 is the fraction of slowly dissolving radionuclide at time, t,

λ_1 is the constant corresponding to the dissolution rate of the rapidly diffusing, soluble fraction into the external solution, and

λ_2 is the constant corresponding to the slow dissolution rate of large particles breaking down.

The first equation describes the removal of the soluble component from the sample by rapid diffusion of small particles into the external solution and its replenishment by the additional fraction as the result of transformation of large particles. The last process is discribed by equation (2).

Let us assume that Q_0 is the initial content of plutonium alpha-activity on the sample filter, S is the transferable portion of radionuclide at the initial moment (the untranferable portion is accordingly 1-S), thus we can write equations describing the initial conditions:

$$Q_1\Big|_{t=0} = S \times Q_0 \qquad (3)$$

$$Q_2\Big|_{t=0} = (1\text{-}S) \times Q_0 \qquad (4)$$

The solution of equations (1) and (2) under initial conditions gives the following expression for initial plutonium content in the sample:

$$Q_0 = Q_1 + Q_2 = Q_0 [a_1 \exp(-\lambda_1 t) + a_2 \exp(-\lambda_2 t)] \qquad (5)$$

$$a_1 = S - (1\text{-}S)\, \lambda_2 / (\lambda_1 - \lambda_2) \qquad (6)$$

$$a_2 = (1\text{-}S)\, \lambda_1 / (\lambda_1 - \lambda_2) \qquad (7)$$

$$S = a_1 + a_2 \lambda_2 / \lambda_1 \qquad (8)$$

Thus equation (8) can be applied to dialysis results to calculate the value, S, the transportable fraction. (S is defined as Transportability). Clearly such an approach to the process kinetics is the rough simplification because polydisperse aerosols, in the early stages of dialysis cannot be described by one term exponentially decreasing with time.

According to equation (8), it is necessary to know all parameters of the two-component exponential model for the determination of transportability, S. These parameters can be obtained only by continuing dialysis for a week or longer, which is not practical. It is possible to simplify the determination S by obtaining the approximate value, S, resulting from two days of dialysis data.

This simplified approach to assessement of transportability is illustrated by the dialysis results from aerosols sampled at different workplaces at the Mayak radiochemical plants. Mean values of the dialysis kinetic parameters obtained by prolonged explorations are given in Table 1. Transportability (S) was calculated with equation (8). Table 1 also contains dialysis data for the first two days (D). The data of Table 1 shows that the alpha activity fraction dialysing in two days, D, coincides with the transportability, S, calculated value. These values for different workplaces range over more than one order of magnitude.

Discussion

The salt composition of Ringer's solution is similar to that of tissues fluids. Dialysis through semi-permeable membranes involving dissolution and diffusion is similar to some mechanisms of lung clearance. It is reasonable to apply the dialysis method to characterization of plutonium aerosols for dosimetry assessment purposes. Our classification results are in good agreement with the ICRP respiratory tract model [1,2]. The dialysis method appears to be a good method for classification of workplace aerosols to predict their clearance times from the respiratory tract.

The clearance time of aerosol particles from the lungs is inversely related to the fraction absorbed into blood. Plutonium oxide aerosols, according to the dialysis

method, are in inhalation class Y with prolonged retention in the alveolar region and poor absorption from the nasal and bronchial regions. Plutonium nitrate and chloride aerosols are in a class with moderate retention in the pulmonary region and moderate absorption from extrathoracic regions. The dialysis data of plutonium citrate, plutonium polymeric nitrate and from aerosol samples collected in different workplaces (see Table 1) indicate the transportability value (S) essentially varies with solubility properties of plutonium compounds. Data in Table 1 shows that the value, S, for aerosols of workplace 3 in the plutonium reprocessing plant (where the plutonium dioxide is the main concern) is more than one order of magnitude lower than that one for aerosols at workplaces 1 and 2 of plutonium reprocessing plant and also at the uranium reprocessing plant (where the mixtures of moderately soluble plutonuium compounds such as nitrate, chloride, and oxalate are present). According to the ICRP lung model for the indicated plutonium compounds, the absorption fractions in the respiratory tract vary within approximately the same ranges. By means of the dialysis method, it is possible to characterize aerosols with respect to their predicted transfer processes in the lung. This approach can be especially useful in the case of workers exposed to mixed plutonium-containing aerosols when the classification by ICRP lung model is not practical [12].

References

1. Annals of the ICRP,Publ.[130:Limits for intakes of Radionuclides by Workers, Pergamon Press, Oxford, 1978.
2. Annals of the ICRP,Publ.[166: Human Respiratory Tract Model for Radiological Protection Pergamon Press, Oxford, 1993.
3. H.Smith,G.N.Stradling,B.W.Loveless and G.J.Ham, Health Phys., 33 (1977) 539.
4. G.M.Kanapilly, O.G.Raabe, C.H.Goh and R.A.Chimenti, Health Phys., 24 (1973) 497.
5. O.G.Raabe, G.M.Kanapily, H.A.Boyd, Inhalation Toxicology Research Institute Annual Report 1972-1973, Albuquerque,1973.
6. A.N.Efimova, A.M.Vorobyev, Gigiena and Canitariya, 1 (1977) 11.
7. V.D.Vashi, C.S.Suryanarayanan, P.Kotrappa, Health Phys.,39 (1980) 108.
8. J.J.Miglio, B.A.Muggenburg, A.L.Brooks, Health Phys.,33 (1977) 449.
9. O.G.Raabe, S.V.Teague, N.L.Richardson, L.S.Nelson, Health Phys., 35 (1978) 663.
10. A.F.Eidson,J.A.Mewhinney, Health Phys., 45 (1983) 1023.
11. K.G. Suslova, R.E. Filipy, V.F. Khokhryakov, S.A. Romanov, R.L. Kathren, Radiation Protection Dosimetry, 67 (1996) 13.

12. V.F.Khokhrykov, S.A.Romanov, K.G.Suslova, Health Effects of Internally Deposited Radionuclides: Emphasis on Radium and Thorium, World Scientific, London (1995) 117.

Table 1. Transportability (S) and the fraction of plutonium alpha activity dialysing in two days (D), from industrial aerosols different radiochemical production workplaces.

Technological proñess step	**Dialysis parameters**		**S,%**	**D,%**	**D / S**
	à, %	**λ, day^{-1}**			
Reprocessing of uranium fuel	a_1 = 2,7 a_2 =97,3	λ_1=1,0 λ_2=0,0023	2,92	2,80	0,96
The same	a_1 = 2,4 a_2 =97,6	λ_1=2,1 λ_2=0,0021	2,50	2,30	0,92
Reprocessing of plutonium fuel, workplace 1	a_1 = 2,44 a_2 = 7,56	λ_1=1,66 λ_2=0,0027	2,60	2,61	1,00
The same, workplace 2	a_1 = 0,84 a_2 =99,16	λ_1=1,10 λ_2=0,00077	0,90	0,80	0,89
The same, workplace 3	a_1 = 0,12 a_2 =99,88	λ_1=0,92 λ_2=0,00025	0,15	0,16	1,07
Average ± S.D.	–	–	–	–	0,97±0,07

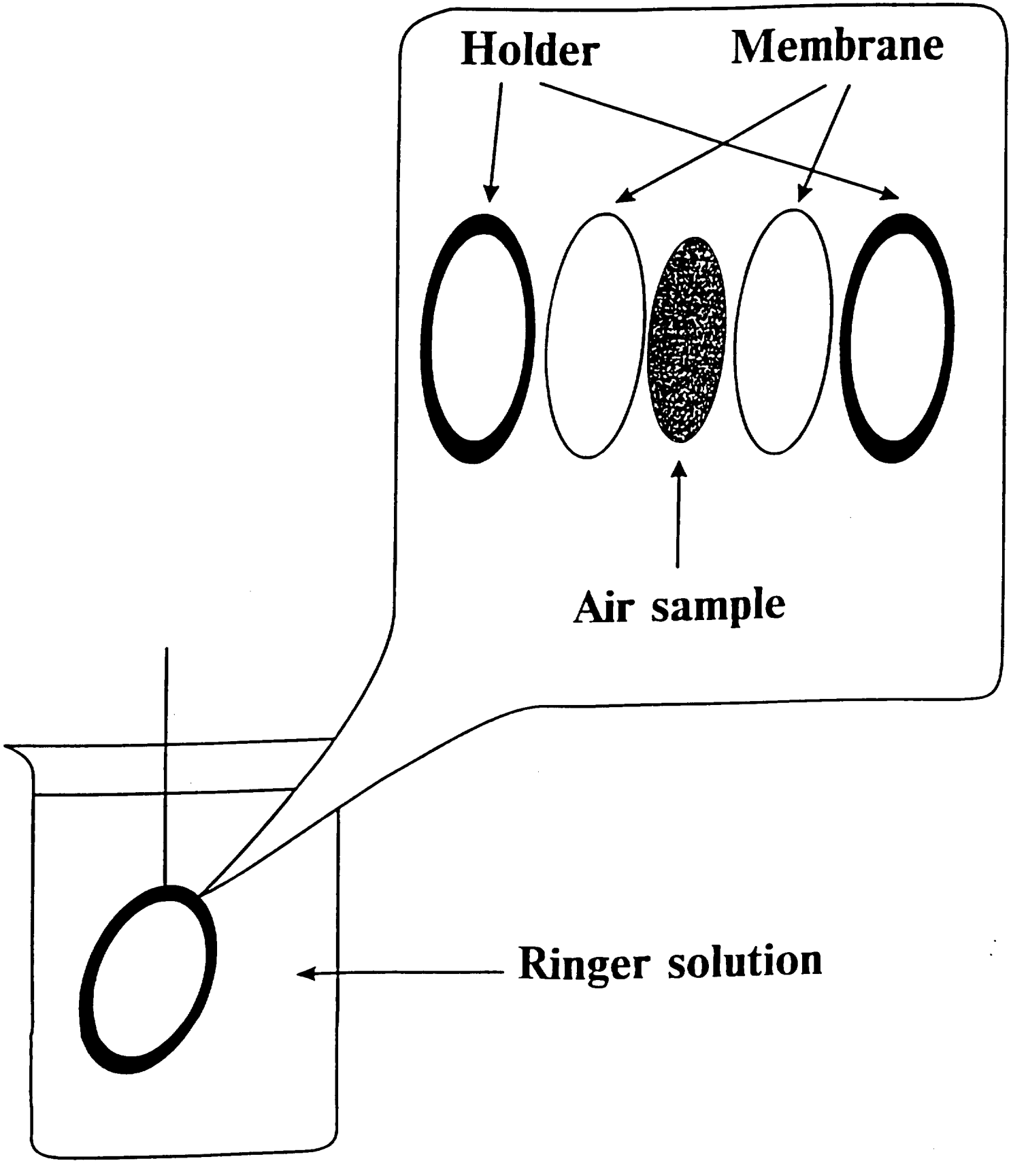
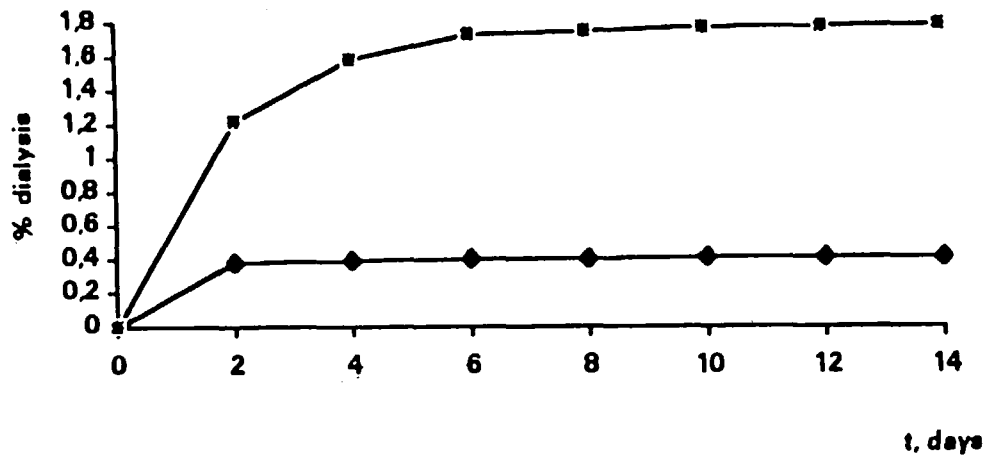

Holder
Membrane
Air sample
Ringer solution

Figure Legends

Figure 1. Assembly of the components for the dialysis method.

Figure 2. The cumulative percentage of plutonium dialysis from aerosols sampled at two different workplaces of the radiochemical plant.


% dialysis
1,8
1,6
1,4
1,2
1
0,8
0,6
0,4
0,2
0
0
2
4
6
8
10
12
14
t, days

A Scintillation method for determination of actinide alpha activity in samples

V. F. Khokhryakov[1], T. I. Kudravtseva[1], V. I. Chernikov[1], K. G. Suslova[1], I. A. Orlova[1], R. E. Filipy[2]

[1]Branch No. 1, Federal Research Center Biophysics Institute, Ozyorsk Road 19, Ozyorsk, Chelyabinsk Region, Russia 456780.

[2]U. S. Transuranium and Uranium Registries, Washington State University, 100 Sprout Road, Richland, WA 99352.

Corresponding Author: R. E. Filipy

ABSTRACT

This report describes an efficient, easy to use, and inexpensive method for detection of alpha activity in biological samples, the alpha radiometer. The actinide elements are coprecipitated from acid-dissovled, ashed samples with bismuth phosphate and the precipitate is mixed with ZnS(Ag) scinitllation powder. The mixture is dried, in a thin layer, on the surface of a polystyrene cuvet and scintillations from the layer are detected with a photomultiplier tube. An optimal counting efficiency is obtained with a scintillation powder thickness between 24 and 37 mg cm^{-2}.

INTRODUCTION

Alpha spectrometry is generally used in radiobiology and radioecology applications for determination of alpha activity in samples. The method has high sensitivity and high resolution; however, its application to large numbers of samples requires expensive equipment. For routine analyses of samples with known isotopic composition, the determination of alpha activities is simpler and less expensive with the alpha radiometer. This has been a useful tool at the First Branch of the Biophysics Institute (BIB-1) for measuring the plutonium content in urine bioassay samples of occupationally-exposed workers of the Mayak plutonium production facility. The device was developed and fabricated over a 20-y period at BIB-1 and it has been in use since 1974. It was certified by the General State Center for Measurement Standards, D. Mendeleev NPO VNIIM, Saint Petersburg, Russian Federation[1]. The purpose of this report is to describe the equipment, the preparation of the samples, and the performance characteristics of the alpha radiometer.

SAMPLE PREPARATION

The radiochemical separation of actinides from the sample were described in previous reports[2,3]. The actinides (plutonium and americium) are separated from the acid-dissolved, ashed urine sample by bismuth phosphate co-precipitation. A solution of bismuth nitrate

and sodium dihydrogen orthophosphate (1:6 by volume) is added to the sample in 1M nitric acid. The actinide-bismuth phosphate precipitate is separated by centrifugation and mixed with ZnS(Ag) scintillation powder and the mixture is suspended in 96% ethanol. The suspension is transferred to a shallow, 25mm-diameter polystyrene cuvet and dried at room temperature, forming a thin layer of ZnS(Ag)-precipitate on the surface of the cuvet. The cuvet is coupled with a low-background Фэу-35 photomultiplier which detects scintillations in the mixture and impulses from the photomultiplier are recorded by a counter (Fig. 1). Up to 20 identical modules are operated simultaneously at the BIB-1 laboratories.

PERFORMANCE

Metralogical characteristics of the low-background alpha radiometer are listed below.

- Range of measurement — 0.001 - 0.5 Bq
- Background level — 0.0009 s^{-1}
- Detector efficiency — 95 $\pm$ 5 %
- Minimum detectable activity (MDA) — 0.001 Bq
- Maximum error of measurement — $\pm$ 18 %

The quantity of the scintillation powder is an important factor influencing the sensitivity of the method. To determine the optimum quantity of ZnS(Ag), standard radioactive solutions of plutonium in nitric acid were added control urine at two activity levels, 0.83 and 2.7 Bq. The urine was processed through the bismuth phosphate coprecipitation and equal aliquots of the precipitate were mixed with varying amounts of scinitllation powder. The mixtures were counted with the alpha radiometer and the results are shown in Table 1, below.

The results in Table 1 show that the highest counting efficiency was achieved when the thickness of the scintillation powder and the precipitate was between 24 and 37 mg cm^{-2}. The mass of ZnS(Ag) corresponding to this thickness was adopted as the optimum for cuvets of different sizes. For routine measurements with the 25mm-diameter cuvet, the optimum mass of scintillation powder is 150 mg. The use of the small cuvet for routine samples has made it possible to decrease both the background of the radiometer and the dimensions of each individual module which is an important consideration when multiple modules are constructed into one unit.

SUMMARY

The alpha radiometer, described above, has been used to analyze the actinide contents in the excreta of more than 7000 personnel of the Mayak plutonium production facility and

in the several tens of thousands of tissue samples collected at autopsy of facility personnel and members of the general population. The method is efficient, easy to use and is reletively inexpensive. The information gained with this method was of great importance to the Mayak dosimetry control system and was used to monitor population exposures to the actinide elements.

REFERENCES

1. Russian Federation State Standard Number 397/96. Radiometer of Alpha Radiation RIA-05, General State Center for Measurement Standards, D. Mendeleev NPO VNIIM, Saint Petersburg, Russia, 1996. (in Russian)

2. Russian Federation State Standard Number 39-2/93. Plutonium: Method for Anion-exchange Estimation of Low-level Alpha Activity in Autopsy Specimens, General State Center for Measurement Standards, D. Mendeleev NPO VNIIM, Saint Petersburg, Russia, 1993. (in Russian)

3. K. G. Suslova, R. E. Filipy, V. F. Khokhryakov, S. A. Romanov, R. L. Kathren, Radiat. Protect. Dosimetry, 67 (1996) 13.

Table 1. Count rate measured by the alpha radiometer with varying thickness of ZnS(Ag) scintillation powder layer in the cuvet.

Thickness of ZnS(Ag) mm cm^{-2}	2.0	6.11	12.2	18.3	24.4	30.5	36.7	42.8	48.9	55.0	61.1
Counting Rate (s^{-1})											
Sample A1[a]	0.08	0.23	0.58	0.69	0.78	0.79	0.78	0.75	0.71	0.65	0.60
Sample A2[b]	0.24	0.75	1.73	2.20	2.45	2.44	2.47	2.43	2.38	2.32	2.20

[a] 0.83 Bq per sample

[b] 2.70 Bq per sample

Figure Legends

Figure 1. Orientation of the photomultiplier with respect to the cuvet containing the actinide precipate-scintillation powder mixture.

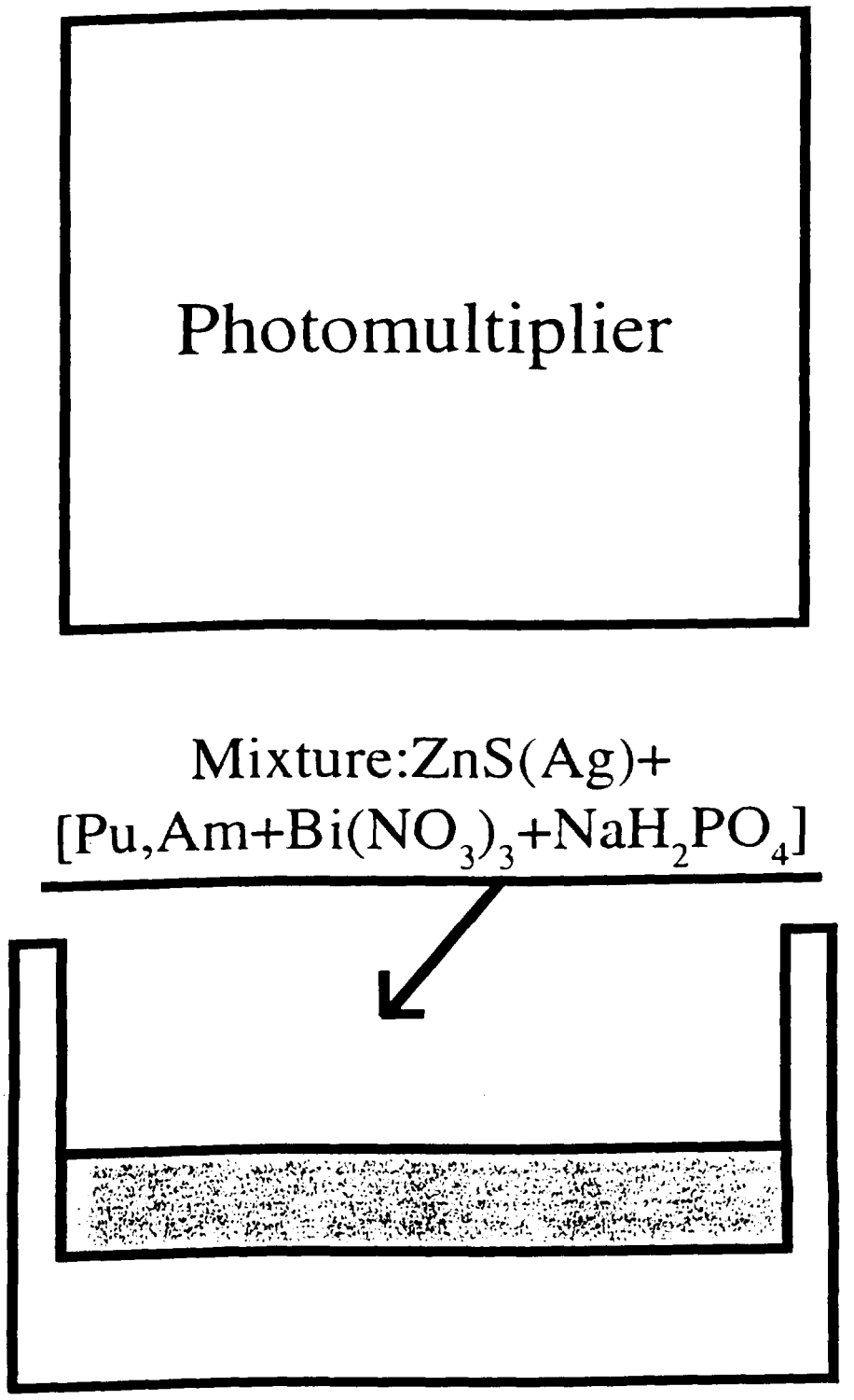
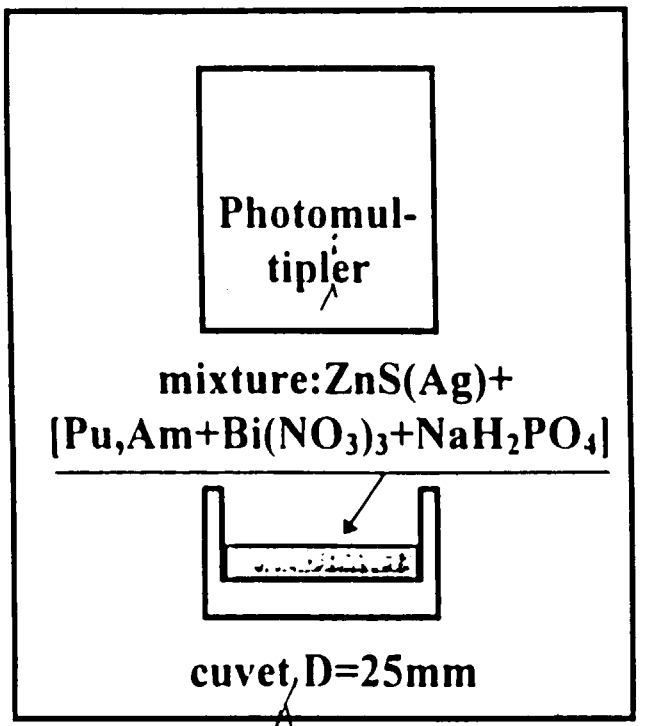

Photomultiplier
Mixture:ZnS(Ag)+
[Pu,Am+Bi(NO3)3+NaH2PO4]
cuvet, D=25mm

Fig. 1


Photomul-
tipler
mixture:ZnS(Ag)+
[Pu,Am+Bi(NO3)3+NaH2PO4]
cuvet, D=25mm

ANALYSIS FOR ACTINIDES IN TISSUE SAMPLES FROM PLUTONIUM WORKERS OF TWO COUNTRIES.

R. E. Filipy*[1], V. F. Khokhryakov[2], K. G. Suslova[2], S. A. Romanov[2], D. B. Stuit[3], E. E. Aladova[2], and R. L. Kathren[1].

[1]U. S. Transuranium and Uranium Registries, Washington State University, Tri-Cities, 100 Sprout Road, Richland, WA 99352, USA.

[2]Branch No. 1 of the Institute of Biophysics, Ozersk Road 19, Ozersk, Chelyabinsk Region, 456780, Russia

[3]U. S. Transuranium and Uranium Registries, Nuclear Radiation Center, Washington State University, Pullman, WA 99163, USA.

Corresponding author: R. E. Filipy

ABSTRACT

For more than 25 years, the United States Transuranium and Uranium Registries (USTUR) and the Dosimetry Registry of the Mayak Industrial Association (DRMIA) of the Russian Federation have, each independently, collected tissues at autopsy from workers with potential or confirmed body burdens of actinide elements resulting from occupational exposures. Tissues, thus obtained, were radiochemically analyzed for actinides for the purpose of evaluating the biokinetics of these elements in the human

body. Scientists of these two organizations have recently begun a collaborative research program to compare, combine and analyze the data to verify or refine biokinetic models needed for radiation dosimetry.

INTRODUCTION

The United States Transuranium and Uranium Registries (USTUR) collects tissue samples at autopsy of volunteer donors who have had a demonstrated occupational intake of one or more of the actinide elements. The program was begun in 1968 and, since then, tissues have been collected from nearly 300 autopsies and from 12 whole-body donors. The collected tissues were radiochemically analyzed to determine the actinide contents at death and this information is used to evaluate the deposition and retention of those elements in the body and to compare the organ contents with estimates of the body burdens made during the life of the individual donor. The primary objective of the USTUR is to ensure the adequacy of radiation protection standards and to verify or suggest modification of the models used as a basis for those standards for the actinide elements.[1,2]

The Dosimetry Registry of the Mayak Industrial Association (DRMIA), operated by Branch No. 1 of the Russian Institute of Biophysics, has been in existence for nearly the same period of time with similar operating procedures and with the same primary objective. Both Registries have operated independently of one another until 1994 when an international agreement for a collaborative research program was consummated between the governments of the Russian Federation and the United States. There are a number of

differences in the methods and scopes of operation of the two Registries as well as many similarities. The purpose of this report is to briefly describe some of these similarities and differences which were addressed, in detail, in another report[3] and to provide a summary of the collaborative research program currently being conducted by the USTUR and the DRMIA.

One of the major differences between the two Registries is in the scope of operation. The workers of Mayak plutonium production facility are the primary concern of the DRMIA who perform the internal dose assessment for the workers of that site. This includes urine bioassays for actinides as well as the in-vivo screening (whole body counting) of Mayak workers. The DRMIA database contains dosimetry and medical records on approximately 5500 workers at the Mayak site. The USTUR, on the other hand, has volunteer donors from nearly all U. S. nuclear production sites and, with a few exceptions, performs no urine bioassays or in-vivo detection assays for actinides. They rely on dosimetry and medical records provided by the employers of their worker-registrants.

Table 1 contains information about the numbers of deceased cases and associated actinide body burdens for which data are maintained in the databases of the two Registries. The DRMIA has performed autopsies on approximately three times the number of cases of the USTUR although, not reflected in the table, the USTUR has had 12 whole body donors and the DRMIA has had none. Results of the radiochemical tissue analyses and the case

descriptions and evaluations for six of these cases have been documented in several USTUR publications. The whole body donations have been extremely valuable in relating the actinide contents of the various tissues or organs of the body to one another and the data from them have been used by both Registries for that purpose. The actinide body burdens of DRMIA cases were also much higher than those of the USTUR cases. In a direct comparison, the mean liver concentration of DRMIA cases was nearly 250 times that of USTUR cases.[3]

RADIOCHEMICAL ANALYSIS

A major difference in the operation of the two Registries lies in the specific radiochemical methods used to analyze previously collected tissue samples. The USTUR has had radioanalytical support from four laboratories from the time of its inception as the National Plutonium Registry in 1968. An analytical laboratory at the Rocky Flats Facility (RF) analyzed tissues donated by those individuals who had been employed at that site. Pacific Northwest Laboratories (PNL) analyzed tissues of all other donors. In 1978, a laboratory at Los Alamos National Laboratory (LANL) also began analyzing tissues for the Registries and for several years, all three laboratories served the Registries. PNL analyzed tissues from approximately 30 cases before their analyses stopped in 1978. RF analyzed tissues from 60 cases before 1987 when their analytical function ceased. LANL analyzed tissues from over 100 cases including 6 whole-body donations before the Washington State University (WSU) laboratory took over the analytical function in 1993. Data from the analysis of tissues from approximately 50 cases were provided to the

USTUR by laboratories in Great Britain. Tissues of all the DRMIA cases have been analyzed by the same methods throughout its history and its methods were quite differenct from those of the USTUR laboratories whose methods differed somewhat over the years.

The DRMIA methods as well as those of all USTUR laboratories began with variations of ashing techniques (wet or dry ashing) and subsequent dissolution of the ashed material in nitric acid. PNL used lanthanum fluoride co-precipitation followed by extraction of plutonium with thenoyltrifluoroacetone (TTA) while RF, LANL, WSU, and the DRMIA laboratories used anion-exchange chromotography to extract the actinides from the acid solution. DRMIA used diethyl-hexyl phosphoric acid (HDEHP) in toluene to separate americium from plutonium while RF, LANL, and WSU used dibutyl-N, N-diethylcarbamyl phosphonate (DDCP) for that purpose. The American laboratories other than PNL used isotopic tracers to quatitate recoveries from the solutions the DRMIA use of tracers was intermittant because of difficulty in obtaining tracers of sufficient purity. For detection of the actinides in samples, PNL used two different methods, both of which were performed on the actinides after electrodeposition on stainless steel disks. An autoradiographic technique[4] was used for low-level samples while direct, electronic counting of alpha particles was used for samples containing a sufficient amount of activity. RF, LANL, and WSU used electrodeposition on stainless steel disks and solid-state alpha detectors. The DRMIA used bismuth phosphate co-precipitation and a Zinc sulfide scintillation method[5] for their higher level samples and, for low-level samples they used the bismuth phosphate

co-precipatation, followed by eletrodepositon and an ion chamber spectrometer. The WSU laboratories followed the procedures developed at LANL[6] for the sake of continuity because LANL analyzed the greatest number of USTUR tissue samples. New, more efficient methods are being investigated by WSU and those methods, after appropriate verification, will likely be used with future samples of WSU and the DRMIA.

DATA COMPARISON

In spite of the differences in radiochemical analytical methods used by the two Registries over the years and regardless of the large difference in actinide body burdens between the two Registries, the resultant data are similar in many respects. In the inital stages of collaboration by the USTUR and the DRMIA, data were compared to determine whether or not they could be combined to evaluate biokinetic models. The first comparison of data involved the use of skeleton-to-liver concentration ratios. The skeleton and the liver are the two major deposition sites in human (or animal) bodies. Ratios were used because they were not expected to differ substantially with high or low body burdens and because estimates of initial depositions and the times of exposure in human subjects are frequently misleading, precluding the use of retention functions for individual organs that are based on initial uptake.

Figure 1 is a plot of skeleton:liver concentration ratios of $^{239+240}$Pu as a function of residence times (the time between exposure or potential exposure and death) and Table 2

contains selected parameters describing those plots. Although the concentration ratios ranged over two orders of magnitude, geometric mean ratios of the two Registries (Table 2) were not significantly different from one another ($P < 0.05$). The ratios of each Registry as well as those of the combined data were log-normally distributed. Slopes of the individual regression lines for the data of each Registry (Figure 1) were not significantly different from one another and were not significantly different from zero ($p < 0.05$); however the slope of the regression line for the combined data was significantly different from zero ($p = 0.03$). Means of ^{241}Am skeleton:liver concentration ratios or the two Registries (Table 2) were also not significantly different from one another ($P < 0.05$). Slopes of the ratio vs residence time regression lines (Figure 2) were also not significantly different from one another and the slopes, including that of the regression of combined data, were not significantly different from zero ($P < 0.05$).

The main difference between the data points of the two Registries was in the residence time parameter. Residence times of the DRMIA cases were generally longer than those of the USTUR cases (Figures 1 and 2) because of the method of calculation. USTUR residence times were the time period between exposure (or potential exposure) and death of the worker and, if no exposure situation was evident, the time period including 67 percent of the worker's time working in a potential exposure situation plus the time

between cessation of work and death[7]. The DRMIA includes the entire period of occupational exposure to plutonium plus the time between cessation of work and death in their residence times. This difference would not be likely to have an appreciable effect on the regressions in Figures 1 and 2; however, the data will be reviewed in the interest of uniformity before they are used in biokinetic models.

FUTURE COLLABORATIVE RESEARCH

A three-year collaborative research project was proposed by the DRMIA and the USTUR and was approved for funding by the U. S. Department of Energy Office of International Health Studies. Twelve major tasks were proposed and the following is a summary of those tasks.

The first task involves intercomparisons of radiochemical analytical methods currently in use by both Registries including a series of performance evaluations with split samples from both laboratories and, ultimately, with standard reference materials prepared by the U. S. National Institute of Standards and Technology. Information thus gained will be used to identify and adjust for consistant differences that might be present in previously collected data of the two Registries before they are combined. Other tasks include establishment of uniform analytical methods to be used by both laboratories for future analyses with respect to sampling methods, ashing methods, actinide separation techniques, spectroscopy methods, and data recording.

Methods used by the two Registries for in-vivo estimates of actinide body burdens will also be compared with the goals of metanalysis of previously collected data and establishment of uniform methods for future collection of data. This task includes calibration of the in-vivo detection equipment used by the DRMIA against that in use in the United States by the exchange of whole-body phantoms. Urinalysis for actinide elements is a tool that has been used by the DRMIA and by the employers of USTUR Registrants to estimate intakes of those elements. The radioanalytical methods used for those estimates will be compared to determine compatibility of the data collected by both Registries.

Radiation dosimetry from plutonium and americium has been a primary goal of both Registries since their inceptions. A number of tasks will be performed in which biokinetics of these elements in occupationally-exposed humans will be examined with DRMIA and USTUR data. Tasks include evaluation of the relationships of actinide concentrations in the lungs, thoracic lymph nodes, and the individual systemic organs at the time of death and comparison of those data with the estimates of body burdens made while the individuals were alive. The purpose of this work is to verify or suggest modifications of models describing the translocation of actinides in the body as proposed by the International Commission on Radiological Protection[8,9].

There are a number of advantages to be gained by collaboration of the two Registries. Collaboration would increase the number of cases available for study by a factor of four for already deceased registrants relative to the number of USTUR cases. Also, there were many more female plutonium workers in Russia than in the United States. The USTUR database contains data from only a few females; therefore, combination of data would result in a greater heterogeneity of the worker population. Because actinide deposition levels in past Russian workers were much higher than those of U. S. workers, dose-dependence or dose-independence of biokinetic parameters should become apparent with the combined databases.

An important part of this work includes the dissemination of information to the world scientific community and the general public. This will be accomplished by prompt, joint publication of the results of the collaborative research in the scientific literature. Also as part of this collaboration, a number of important Russian documents regarding plutonium metabolism and dosimetry, previously classified, will be translated to English and distributed through the scientific literature.

The dosimetric information resulting from this project will be in direct support to other projects which are a part of the overall program. The objectives of two other, separate projects are to relate internal doses combined with external doses to stochastic effects, such as cancer, and to deterministic effects, such as blood dyscrasia, noted in workers at

the Mayak plutonium production site. Still other projects of the joint U. S.- Russian program are concerned with dosimetry as well as stochastic and deterministic effects in the general Russian population residing in the vicinity of the Mayak site and the results of this worker dosimetry project are expected to be helpful in the dose reconstruction efforts for that population. The biokinetic data and organ doses, in conjunction with biological effects, will be used to develop and define risk coefficients for those effects resulting from chronic exposure to relatively high levels of radiation.

REFERENCES

1. R. L. Kathren; Radiat. Protect. Dosimetry, 26 (1989) 323.

2. R. L. Kathren, Radiat. Protect. Dosimetry, 53 (1994) 1.

3. K. G. Suslova, R. E. Filipy, V. F. Khokhryakov, S. A. Romanov, R. L. Kathren, Radiat. Protect. Dosimetry, 67 (1996) 13.

4. L. C. Schwendimen and J. W. Healy, Nucleonics, 16 (1958) 78.

5. V. F. Khokhryakov, T. I. Kudryavtzeva, V. I. Chernikov, I. A. Orlova, This volume.

6. J. F. McInroy, H. A. Boyd, B. C. Eutsler, D. Romero, Health Phys. 49 (1985) 587.

7. R. E. Filipy, R. L. Kathren, Health Phys. 70 (1996) 153.

8. International Commission on Radiological Protection, Human Respiratory Tract Model for Radiological Protection, Publication 66 (Oxford: Pergamon Press) Ann ICRP 24(3/4) 1994.

9. International Commission on Radiological Protection, Age-dependent Doses to Members of the Public from Intake of Radionuclides, Part 2, Publication 67 (Oxford: Pergamon Press) Ann. ICRP 23(3/4), 1993.

Table 1. A comparison of the USTUR and DRMIA databases.

Registry	Nuclide	Program Started	Total Number of Autopsies	Range of Body Burdens
DRMIA	$^{239+240}Pu$	1965	870	40 Bq - 175 kBq
	^{241}Am	1975	460	2.0 Bq - 4.5 kBq
USTUR	$^{239+240}Pu$	1968	280	40 - 300 Bq*
	^{241}Am	1968	280	1.0 - 150 Bq*

*A general range with a few lower and a few higher burdens.

Table 2. Selected parameters describing the skeleton:liver concentration ratios for plutonium and americium in the bodies of workers of the United States and the Russian Federation.

	USTUR	DRMIA	Combined data
$^{239+240}Pu$			
No. of Cases	66	74	137
Geometric Mean[a]	-0.81	-0.64	-0.71
GSD[b]	0.37	0.22	0.27
Slope[c]	-0.0020	0.0010	0.0060
P[d]	0.77	0.92	0.028
^{241}Am			
No. of Cases	30	42	74
Geometric Mean	-0.32	0.62	-0.064
GSD	0.35	0.36	0.45
Slope	0.0040	-0.0040	0.015
P	0.59	0.63	0.29

[a] Mean of the logarithms of skeleton:liver concentration ratios

[b] Geometric standard deviation of the concentration ratios

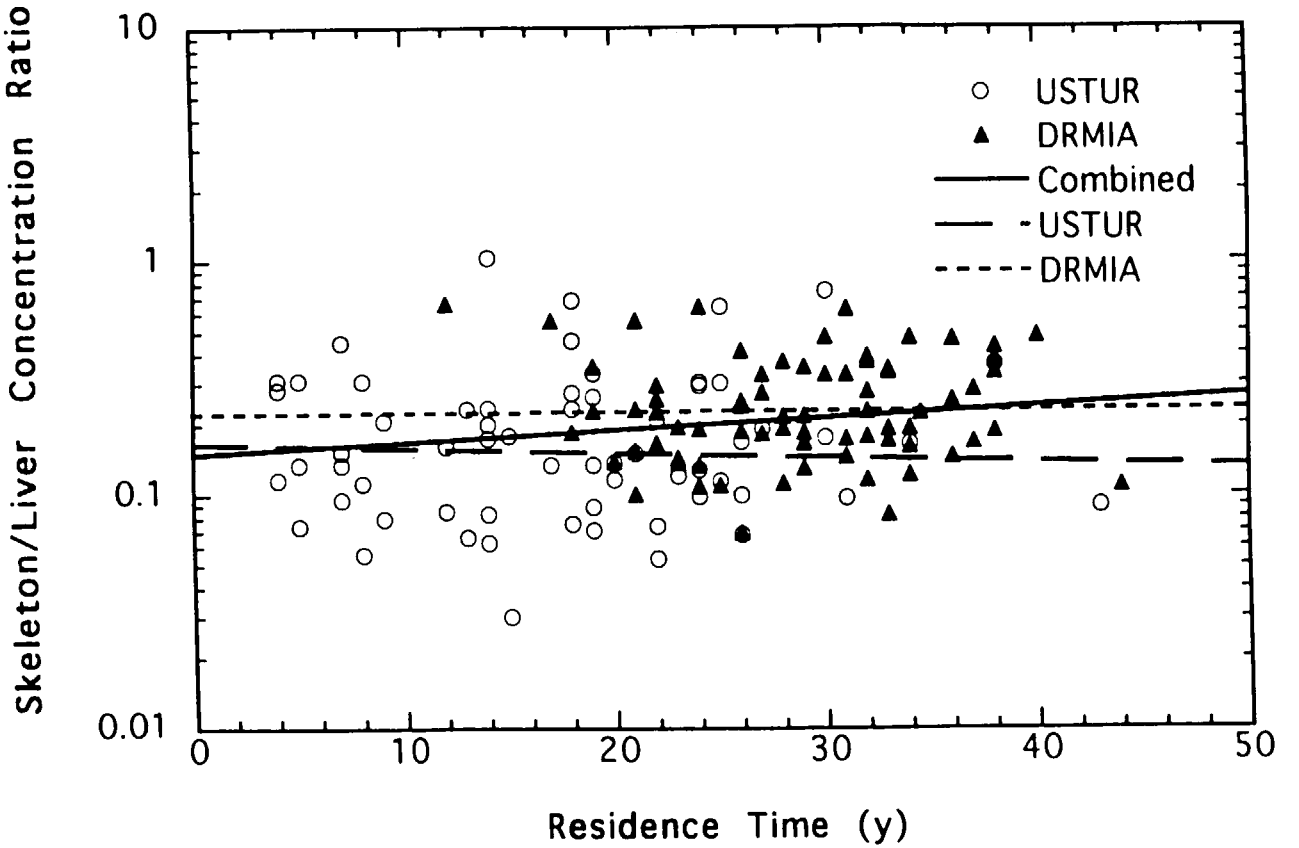
[c] Slope of the regression lines relating the logarithms of concentration ratios to residence times (time between exposure and death).

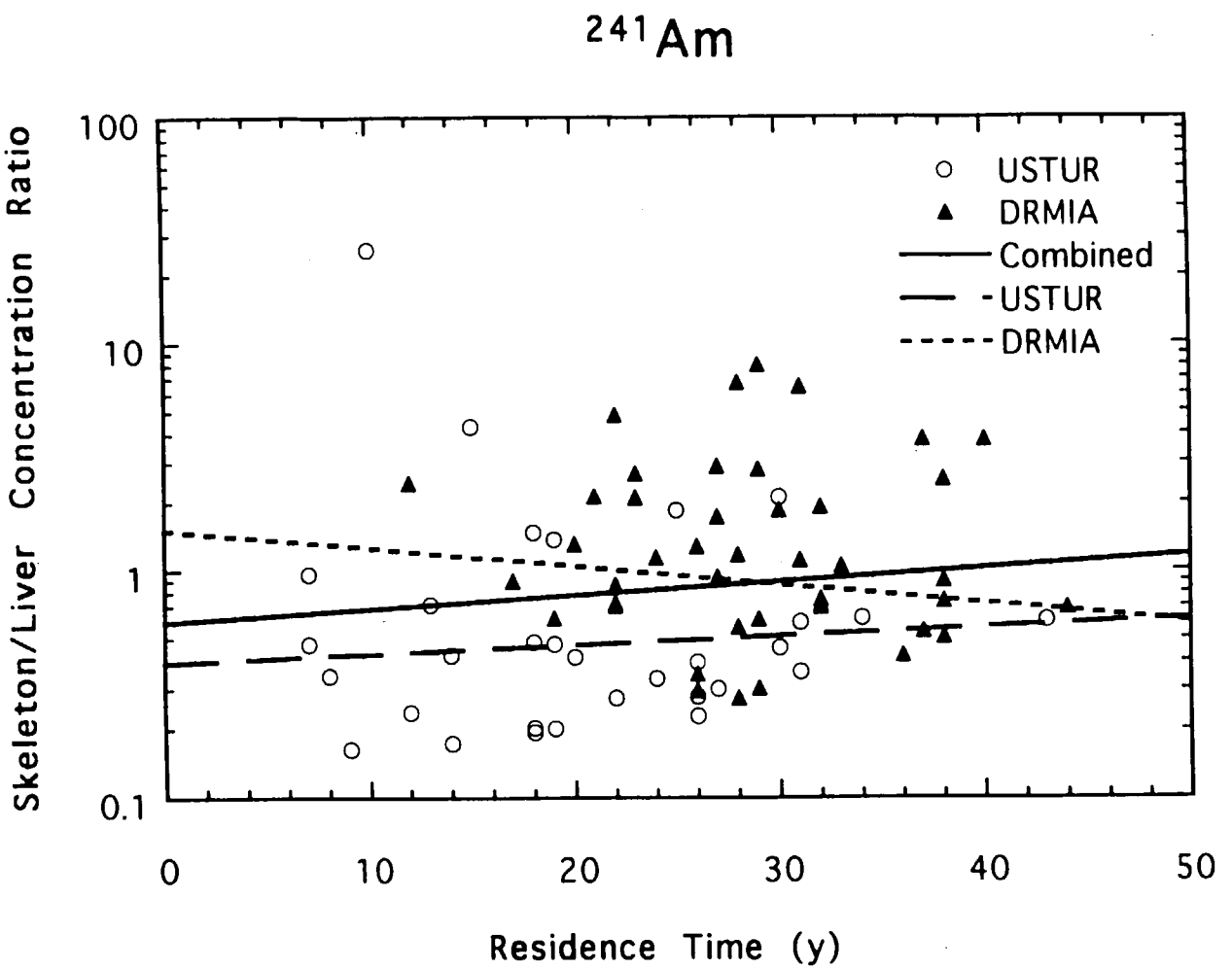
[d] Probability that the slope of the regression line was not significantly different from zero ($P < 0.05$).

Figure Legends

Figure 1. Skeleton-to-liver ^{241}Am concentration ratios as a function of residence time (time between exposure and death) in workers occupationally exposed to actinide elements.

Figure 2. Skeleton-to-liver ^{239}Pu concentration ratios as a function of residence time (time between exposure and death) in workers occupationally exposed to actinide elements.


239Pu
Skeleton/Liver Concentration Ratio
10
1
0.1
0.01
0
10
20
30
40
50
Residence Time (y)
USTUR
DRMIA
Combined
USTUR
DRMIA


241Am
Skeleton/Liver Concentration Ratio
100
10
1
0.1
0
10
20
30
40
50
Residence Time (y)
USTUR
DRMIA
Combined
USTUR
DRMIA